SUB-ÅNGSTROM TRANSMISSION ELECTRON MICROSCOPY FOR MATERIALS SCIENCE

Michael A. O'Keefe, E.C. Nelson and Ulrich Dahmen

National Center for Electron Microscopy, Materials Science Division, Ernest Orlando Lawrence Berkeley National Laboratory, Berkeley, CA94720, U.S.A.

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof, or The Regents of the University of California.

Ernest Orlando Lawrence Berkeley National Laboratory is an equal opportunity employer.

Ernest Orlando Lawrence Berkeley National Laboratory – LBNL-48466

SUB-ÅNGSTROM TRANSMISSION ELECTRON MICROSCOPY FOR MATERIALS SCIENCE

Michael A. O'KEEFE, E.C. NELSON and Ulrich DAHMEN

National Center for Electron Microscopy, Materials Science Division, Ernest Orlando Lawrence Berkeley National Laboratory, Berkeley, CA94720, U.S.A.

High-resolution electron microscopy (HREM) is becoming even more important as materials scientists build artificially-structured nano-materials. Nanostructures often include atoms with bond lengths shorter in projection than the point resolution of a standard mid-voltage HREM[1]. Image simulations show that structure determination of defects such as dislocation cores need sub-Ångstrom resolution[2]. Sub-Angstrom TEM is also important for atomic-level analysis of interface structure and for tomographic reconstruction of 3-dimensional shapes of nanocrystals. Research on embedded nanocrystals show that this knowledge is essential for understanding of magic-size behavior recently found for Pb inclusions in Al[3]. Similar needs arise in the characterization of nanoscale dynamic phenomena such as size-dependent phase separation, superheating or premelting in small particles or inclusions.

The one-Ångstrom microscope (OÅM) project[4] at the National Center for Electron Microscopy has demonstrated sub-Ångstrom TEM to a resolution of 0.78Å. By combining a modified CM300FEG-UT with computer software[5,6], we are able to generate sub-Ångstrom super-resolution images from experimental image series.[7]

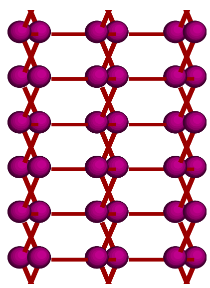
Sub-Ångstrom microscopy with a high-resolution transmission electron microscope requires meticulous attention to detail. As resolution is improved, resolution-limiting parameters need to be reduced. In particular, aberrations must be minimized, power supplies must be stabilized, and the microscope environment optimized to reduce vibration and acoustic and electromagnetic noise. For a direct resolution of d_S the spherical aberration coefficient C_S needs to be below $6d_S^4/\lambda^3$. To reach 0.8Å directly at 300keV would require C_S to be less than 0.03mm (0.02mm would be optimum[8]).

Alternatively, to resolve an information limit of d_Δ by focal reconstruction requires the microscope's standard deviation of focus spread Δ to be less than $2d_\Delta^2/(\pi\lambda)$, or 21Å for d_Δ=0.8Å at 300keV. Two- and three-fold astigmatism, A_1 and A_2 must also be small. To ensure phase distortions of less than $\pi/4$ at resolutions of d_{A_1} and d_{A_2}, values of A_1 and A_2 must be below $d_{A_1}^2/(4\lambda)$ and $3d_{A_2}^3/(8\lambda^2)$ respectively, or 8Å and 500Å to reach 0.8Å at 300keV. In addition, specimen thickness must be kept to less than $2d_S^2/\lambda$, requiring specimens thinner than 65Å to reach 0.8Å at 300keV.[9]

We achieved sub-Ångstrom resolution with the OÅM by extending its information limit d_Δ with improved high-voltage and lens-current power supplies, and placing it in surroundings specially-constructed to minimize noise[10]. With a hardware corrector, we reduced the OÅM's three-fold astigmatism A_2 from 2.46µm to 30nm so as to extend its $\pi/4$ phase limit to 0.68Å and allow the OÅM to image diffracted beams out to its information limit without introducing significant three-fold distortion.[1]

Measurement of the energy spread (gun spread plus high-voltage ripple) as 0.93eV FWHH indicated a focus spread Δ of 20Å and an information limit of 0.78Å. Tests with a diamond specimen showed that A_2 was corrected and the OÅM could successfully resolve the 0.89Å (400) dumbbell spacings in [110] diamond.[1, 7, 11, 12]

To explore smaller spacings, we lowered the energy spread of the OÅM electron gun to reduce spread of focus from 20Å to 18Å, and thus produce an information limit of 0.75Å. Using a silicon specimen tilted into [112] orientation and imaging at the 0.78Å alpha-null defocus[13], we were able to transfer 444 diffracted beams into the image, and generate images showing atoms as white spots at 0.78Å separation.



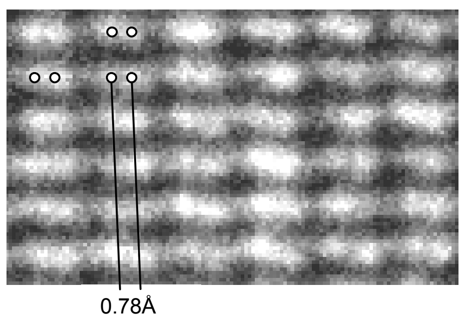


Figure 1. Atom model (left) and OÅM image (right) of silicon in [112] orientation both show the 0.78Å separation (marked) of atom pairs. Three of the atom pairs in the OÅM image are marked with black circles to indicate the atom positions.

REFERENCES

1. Y.C. Wang et al, in *57th Ann. Proc. MSA,* Portland, Oregon 822-823 (1999).
2. C. Kisielowski et al., in *58th Ann. Proc. MSA*, Philadelphia, Penn. 16-17 (2000).
3. U. Dahmen, S.Q. Xiao, S. Paciornik, E. Johnson and A. Johansen, *Phys. Rev. Lett.* 78: 471 (1997)
4. M. A. O'Keefe, in *Microstructures of Materials,* ed. K. Krishnan, 121-126 (1993).
5. W.M.J. Coene, A. Thust, M. Op de Beeck and D. Van Dyck, *Ultramicroscopy* 64: 109-135 (1996).
6. A. Thust, W.M.J. Coene, M. Op de Beeck and D. Van Dyck, *Ultramicroscopy* 64: 211-230 (1996).
7. M.A. O'Keefe, in *58th Ann. Proc. MSA*, Philadelphia, Penn. 1192-1193 (2000).
8. M.A. O'Keefe, in *58th Ann. Proc. MSA*, Philadelphia, Penn. 1036-1037 (2000).
9. M. A. O'Keefe, in *55th Ann. Proc. MSA,* Cleveland, Ohio 1165-1166 (1997).
10. John H. Turner, Michael A. O'Keefe & Robert Mueller, *55th Ann. Proc. MSA,* Cleveland, Ohio 1177-1178 (1997).
11. Michael A. O'Keefe et al., *Ultramicroscopy* (2001) submitted.
12. C. Kisielowski et al., *Ultramicroscopy* (2001) submitted.
13. Michael A. O'Keefe, in *59th Ann. Proc. MSA*, Long Beach, California (2001).

Work supported by the Director, Office of Science, through the Office of Basic Energy Sciences, Material Sciences Division, of the U.S. Department of Energy, under contract No. DE-AC03-76SF00098.